

COMBUSTION SYNTHESIS OF CARBON NANOSTRUCTURES USING ROTATING COUNTERFLOW DIFFUSION FLAMES

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Introduction

A hydrocarbon flame can naturally and readily provide an appropriate high-temperature environment with abundant radical concentrations required for the growth of carbon nanotubes (CNTs) [1] or carbon nano-onions (CNOs) [2]. Therefore, flame synthesis shows a more promising potential for inexpensive and mass production of high-purity carbon nanostructures than other synthesis methods do. Recently, great efforts have been devoted to studying flame synthesis of CNTs. As described in our previous studies [3] rotating flow significantly affected the temperature distribution, mixing of fuel and oxidizer, and residence time of the flow, which in turn were expected to affect greatly the hydrocarbon reactants and environments for the formation of CNTs. However, rotating flow fields have not been applied to control growth of CNOs in flames prior to this study. Therefore, in this paper we aim at exploring the influence of flow rotation on the synthesis of carbon nanomaterials using rotating counterflow diffusion flames and a catalytic Ni substrate.

Experimental

A schematic of the apparatus is shown in Fig. 1. The counterflow system consisted of two identical vertically-aligned cylindrical burners with an inside diameter of 46 mm and an outside diameter of 50 mm, which can rotate about their axis of symmetry. Each burner contained series of small wire-mesh screens and honeycombs to produce uniform velocity profile at the exit plane of burner and to insure rigid-body rotation of the gas inside the burner. The separation distance between the burner exit planes in counterflow, L , was kept at a constant value (22 mm) in the experiment. Oxygen, nitrogen and ethylene were separately metered by conventional rotameters. Then, a stable quasi-one-dimensional diffusion flame was established by introducing two opposite streams of gases in the upper and lower burners; the mixture of ethylene (purity 99.5%) and nitrogen (purity 99.9%) was introduced from the upper burner and the oxidizer (O_2+N_2) was supplied from the lower burner. Throughout each test, the velocities (V) at the exit plane of the upper and lower burners were maintained identical. Also, the angular velocities (ω) of the upper and lower burners had the same magnitude and direction (Fig. 1).

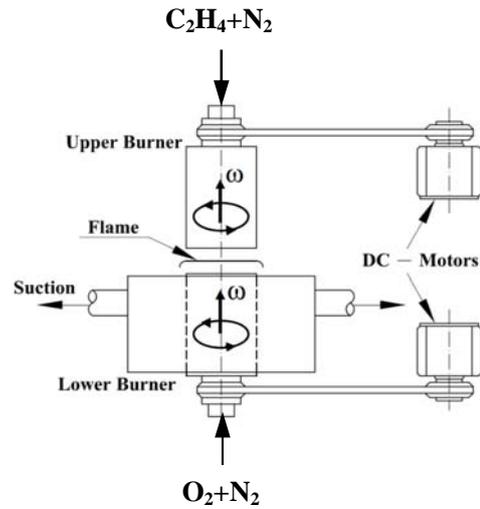


Fig. 1 Schematic of rotating counterflow configuration.

A nascent nickel mesh (200 mesh) with a diameter of 3mm and a thickness of 0.2 mm was used as the catalytic metal substrate to collect deposit materials. During the synthesis of carbon nanostructures, the substrate was positioned at the fuel side near the flame front, denoted by an axial distance z . Here the origin of the cylindrical coordinates (r, z) was located at the center of the lower burner exit. The axial location of the upper edge of the flame front was denoted by z_f . In this study, we used 80% N_2 -diluted C_2H_4 ($\Omega_F=20\%$) as the fuel and 40% $O_2+60\%N_2$ ($\Omega_O=40\%$) as the oxidizer. The deposition time was kept at 120 sec. At the sampling position, the gas temperature ranged from about 600 to 800 °C as determined by a fine wire silica-coated R-type thermocouple (Pt/Pt-13% Rh) with a 0.05 mm diameter. A field emission scanning electron microscopy (FE-SEM) was used to characterize the deposit materials.

Results and Discussion

Fig. 2(a) demonstrates that the diffusion flame was located at the fuel side ($z_f = 11.5$ mm) when $\omega=0$ rps under the condition of $\Omega_O=40\%$, $\Omega_F=20\%$ and $V=15$ cm/s. As ω was increased from $\omega = 0$ to 4 rps, the diffusion flame shifted towards the upper fuel nozzle. The onset of recirculation zone occurred at the angular velocity $\omega = \omega_{cr}$. Here $\omega_{cr} = 3.4$ rps for the flow field with $V = 15$ cm/s and $L = 22$ mm. At $\omega = 5$ rps, the flame resided at a slightly lower axial position than $\omega = 4$ rps because $\omega > \omega_{cr}$. As shown in Fig. 2(b), the gas temperatures at the location of 1 mm above the flame ($z = z_f+1$ mm) were within the range of 600~800 °C for various angular velocities. The temperature first increased and then decreased with increasing ω . At $\omega = 5$ rps, flame temperature was slightly lower than $\omega = 4$ rps because the stretch rate changed from positive to negative. In other words, at $\omega = 5$ rps, the

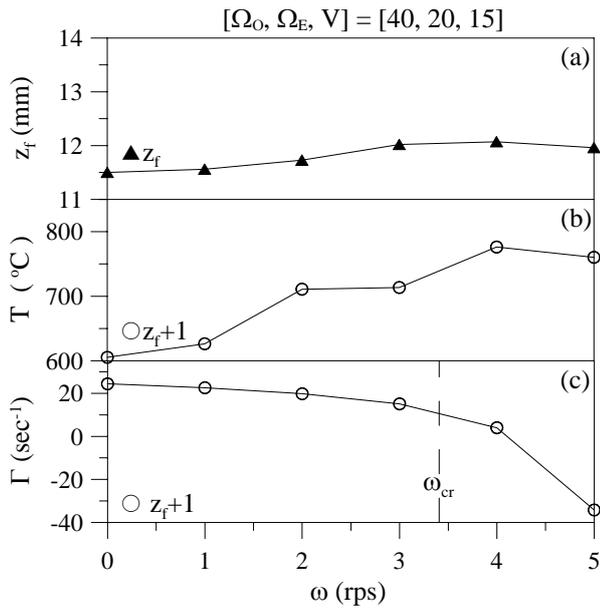


Fig. 2 Variations of (a) flame position, (b) gas temperature and (c) strain rate (Γ) at $z = z_f + 1$ mm.

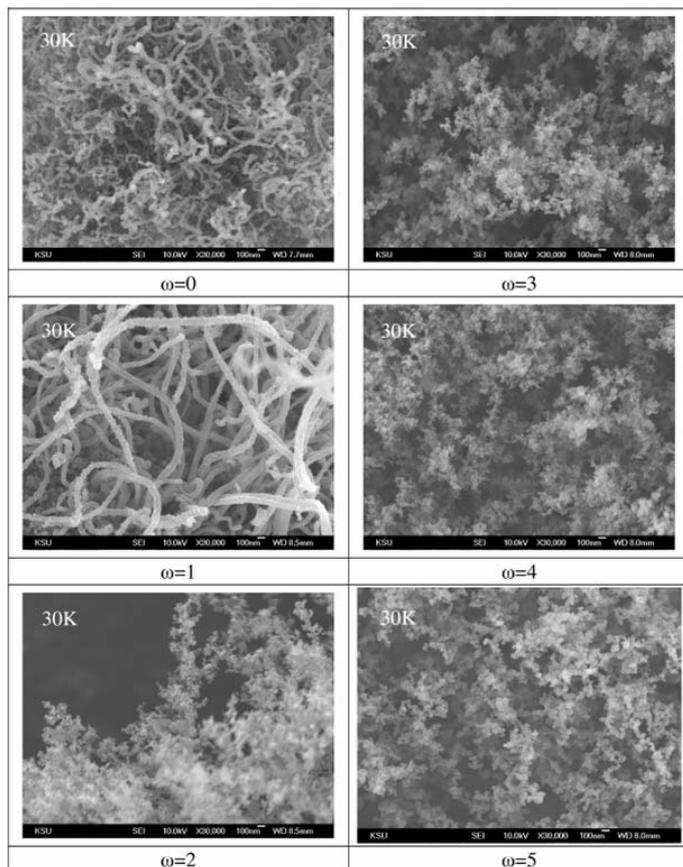


Fig. 3 Typical SEM images of CNTs and CNOs synthesized at $z = z_f + 1$ mm for $\Omega_o = 40\%$, $\Omega_f = 20\%$ and $V = 15$ cm/s.

flame endured compression effect ($\Gamma < 0$). Fig. 2(c) shows that as the angular velocity increased, the stretch rate decreased, leading to the increase of residence time. Evidently, flow rotation lengthened the residence time and resulted in the formation of a soot layer on the fuel side of the diffusion flame. In this study, the process of soot formation was dominated by the angular velocity ω .

It was recognized that stain rate affected carbon nanostructures synthesis either through the residence time of the flow or carbon sources available for carbon nanostructures growth. A diffusion flame at high angular velocity (low strain rate) was stronger than a weak flame at low angular velocity (high strain rate) and produced more carbon sources due to the longer residence time of the flow. Fig. 3 shows the SEM images of CNTs and CNOs influenced by angular velocity (ω). At a higher strain rate ($\omega = 0$ and 1 rps), curved and entangled tubular multi-walled CNTs were formed, however, at a lower strain rate ($\omega = 2 \sim 5$ rps) CNOs were synthesized. Note that the temperature range for the CNOs synthesis was higher than that for CNTs. It was verified that flow rotation associated with residence time played an important role in the synthesis of carbon nanostructures, namely the microstructures and yield.

Conclusions

The influence of flow rotation on the synthesis of carbon nanostructures was investigated using rotating counterflow ethylene diffusion flames and a catalytic Ni substrate. In the experiments, the fuel and oxidizer compositions of 20% $C_2H_2 + 80\% N_2$ and 40% $O_2 + 60\% N_2$ in the upper and lower burners, respectively, were kept constant, whereas the strain rate was varied by adjusting the rotational speed. Stain rate affected carbon nanostructures synthesis either through the residence time of the flow or carbon sources available for CNTs and CNOs growth. A diffusion flame at a low strain rate was stronger than a weak flame at a high strain rate and produced more carbon sources because of the longer residence time of the flow. At a higher strain rate, curved and entangled tubular multi-walled CNTs were formed, however, at a lower strain rate carbon nano-onions (CNOs) were synthesized. It was verified that flow rotation associated with residence time played an important role in the synthesis of carbon nanostructures, namely the microstructures and yield.

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